Radiative lifetimes in Ni I

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New radiative lifetime measurements based on time-resolved laser-induced fluorescence are reported for 57 odd-parity and 9 even-parity levels of Ni I, ranging in energy from 29 000 to 57 000 cm^{-1}. Our lifetimes agree with earlier measurements based on laser-induced fluorescence but are much more extensive than earlier laser measurements. These lifetimes are found to agree with the recent critical compilation of atomic transition probabilities from the U.S. National Bureau of Standards [J. Phys. Chem. Ref. Data **17,** Suppl. 4 (1988)]. These lifetimes are also compared with lifetimes derived from the highly accurate (±0.7%) relative oscillator strengths measured by the Oxford group [Mon. Not. R. Astron. Soc. **236,** 235 (1989)]. Our lifetimes provide an improved (±2%) absolute scale for the Oxford measurements.

INTRODUCTION

A combination of techniques from laser spectroscopy and Fourier transform spectroscopy is rapidly improving the database of atomic transition probabilities. Recent studies of V I,[1] of Sc I and Sc II,[2] and of Fe I (Ref. 3) demonstrate the power of this approach. Laser-induced fluorescence (LIF) from atoms and ions in a beam is used to measure accurate radiative lifetimes. These lifetimes provide the normalization needed to convert branching fractions, as measured with a powerful Fourier transform spectrometer, into absolute transition probabilities. We report accurate (±5% or ±0.2 ns) lifetimes for 66 levels in Ni I, with energies ranging from 29 000 to 57 000 cm^{-1}, measured by time-resolved LIF on a slow Ni atomic beam. The lifetimes provide the absolute normalization needed for comprehensive transition probability measurements of Ni I by Fourier transform spectroscopy. A comprehensive set of Ni I transition probabilities will be useful, for example, in studying departures from local thermodynamic equilibrium in stellar atmospheres.[4] These lifetime measurements confirm and extend earlier measurements based on LIF methods[5,6] and confirm the normalization for Ni I transition probabilities in a recent critical compilation.[7] The lifetimes provide an improved absolute normalization for the highly accurate (±0.7%) relative absorption oscillator strengths from the Oxford group.[4]

EXPERIMENT

O'Brian *et al.*[3] recently described the apparatus used in this experiment. A hollow-cathode beam source is used to produce a slow beam of Ni atoms, a pulsed dye laser is used to excite these atoms, and the resulting fluorescence decay is detected and analyzed to determine a radiative lifetime. No further description will be given here, but a discussion of potential systematic errors is essential.

RANDOM AND SYSTEMATIC UNCERTAINTIES

A conservative estimate of the total random and systematic uncertainty in our lifetimes is the larger of ±5% or ±0.2 ns. The signal from the photomultiplier (PMT) in this experiment is recorded and averaged with a Tektronix SCD1000 transient waveform digitizer, which has an analog bandwidth of 1 GHz and a sampling rate as high as 200 GHz. The leading edge of the signal from the PMT and the first 5 ns after the peak are discarded in our analysis of the LIF. Thus deconvolution of the fluorescence signal and the laser pulse is avoided. The signal is analyzed by using a least-squares fit to a single exponential. Reported lifetimes are an average of more than 12 000 individual fluorescence decay curves, each of which represents as many as 10^4 photons. The statistical uncertainty is less than 0.5% (1 standard deviation) when signals are strong.

The lifetime experiment has a dynamic range of 2 ns to 2 μs. The finite electrical bandwidth of the detection apparatus is a concern at the 2-ns end of the range. Error arising from the atoms escaping from the observation region before radiating is a concern at the 2-μs end of the range.

The base of the 1P28A PMT is wired for low overall inductance in order to maintain the full electronic bandwidth of the tube. The base includes bypass capacitors in order to provide good linearity to 10 mA of peak anode current and includes small damping resistors to reduce ringing.[8] The electrical bandwidth of the detection system was previously tested by measuring the lifetimes of the 3 $^1P_1^\circ$ and 4 $^1P_1^\circ$ levels of He I, which are known to ±1% from sophisticated calculations.[9,10] We also measure the lifetime of the $2s2p$ $^1P_1^\circ$ level of Be I to test the bandwidth of our experiment. Although the calculations available for the $2s2p$ $^1P_1^\circ$–$2s^2$ $^1S_0$ transition probability in Be I are not so accurate as the best calculation for He I, we believe that there is enough evidence to conclude that the radiative lifetime of the $2s2p$ $^1P_1^\circ$ level is 1.85 ± 0.05 ns.[11] The Sims and Whitten[11] calculation of the $2s2p$ $^1P_1^\circ$–$2s^2$ $^1S_0$ transition probability in Be I approaches the reliability of the best calculations for He I. Accurate (±1% to ±3%) experimental lifetimes in the 3–8-ns range are available for selected levels in Fe II and Cu I.[12,13] We remeasure some of these Fe II and Cu I lifetimes to test the experiment in the 3–8-ns range. We measure the lifetime of the 3 $^3P^\circ$ level

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of He I, which is known to ±1% from sophisticated calculations, to test the experiment in the 100-ns range.[10]

Optimum fluorescence collection efficiency for levels with lifetimes of ≤100 ns is achieved by imaging the intersection of the laser and the atomic beams onto the PMT cathode. Two lenses, forming an $f/1$ system with unity magnification, are used. The fluorescence light is roughly collimated between the two lenses. There is a provision for inserting interference filters or dye filters between the lenses. Occasionally, branching ratios are favorable for observing fluorescence at a wavelength much different from the laser wavelength, in which case filters are used to block scattered laser light and isolate the LIF.

Imaging the intersection of the laser and the atomic beams directly onto the PMT cathode is not optimum for longer-lived (≥300 ns) levels, because this arrangement can lead to systematic error in long lifetimes caused by atomic motion. The image of the radiating atoms moves across the photocathode, which has a position-dependent response. In addition, the efficiency of the fluorescence collection system is weakly position dependent. This systematic error is said to be due to the atoms' escaping from the observation region before radiating. This systematic error was studied in detail during earlier experiments with the same apparatus,[14] and techniques were developed for eliminating the error. One of the techniques involves introducing optics into the fluorescence collection system to defocus the image of the atomic beam on the photocathode. (This modification results in loss of fluorescence signal.) The second technique, which is necessary when lifetimes near 2 μs are measured, involves time-of-flight selection of slow atoms. All the Ni I lifetimes are less than 500 ns, thus only the former technique is necessary.

Repopulation of the level under study by radiative cascade from higher levels is not a problem because of the highly selective laser excitation. Filters are used to block emission from lower-lying levels, which are populated by radiative cascade from the upper level under study. Effects of this cascade fluorescence are observed, and are eliminated by filters, for odd-parity levels above 44 000 cm^{-1}, such as levels of the $x\ ^3P°$, $v\ ^3D°$, $^5S°$, and $w\ ^3P°$ terms. These levels have significant branches in the infrared.

Even-parity levels studied here are all excited from the $z\ ^5G^\circ_6$, $z\ ^5D^\circ_4$, and $z\ ^5D^\circ_3$ levels, which are effectively metastable odd-parity levels. Lifetimes measurements on even-parity levels, such as those in the $e\ ^5F$ and $e\ ^3G$ terms, require considerable care in the analysis of possible cascade fluorescence and in the choice of the blocking filter.

The ±5% or ±0.2 ns total uncertainty in our measurements cannot be described simply as a 1σ or 3σ uncertainty, because the statistical uncertainty in our results is quite small (<1%), as described previously. It is our goal to have more than 90% of our lifetimes accurate within one uncertainty. Every effort is made to understand and to control systematic error.

RESULTS

Table 1 is a list of our lifetime measurements. The number in parentheses following each entry is the uncertainty in the last digit of the entry. Configuration and term assignments, along with level energies, are from Ref. 15. Laser wavelengths used to excite the levels are also listed. The wavelengths are from Ref. 16. Whenever possible, more than one transition is used to excite a level. This duplication ensures that the original classification of the line is correct, that the line is unblended, and that it is correctly identified in our experiment.

The critical compilation of atomic transition probabilities by Fuhr *et al.* reviewed research before 1988 on Ni I transition probabilities.[7] Thus the comparison in Table 1 is primarily between the present study and their critical compilation. We also include earlier LIF measurements of lifetimes in Table 1 because they are directly comparable with our measurements. The average and root-mean-squared differences between earlier lifetime measurements by LIF of Heldt *et al.*[6] and our lifetimes are 8.9% and 10.8%, respectively.[6] The average and root-mean-squared differences between earlier lifetime mea-

Table 1. Radiative Lifetimes in Ni I[a]

Configuration and Term		J	Level (cm^{-1})	Laser Wavelength in Air (nm)	Lifetime (ns) This Expt.	Other LIF	Critical Compilation[b]
$3d^8(^3F)4s4p(^3P°)$	$z\ ^5F°$	4	29 084.478	343.728, 346.166	31.1(16)		31.4
		3	29 832.810	345.289, 337.423	71(4)	69(2)[c]	67.3
		2	30 163.140	341.394, 351.395	190(10)		168
$3d^9(^2D)4p$	$z\ ^3P°$	2	28 569.210	352.454, 361.045	8.4(4)		9.1
		1	29 500.690	349.297, 359.770	8.3(4)		8.6
		0	30 192.268	351.034, 691.456	8.1(4)		8.3
$3d^9(^2D)4p$	$z\ ^3F°$	3	29 320.782	351.506, 357.187	12.4(6)		13.9
		4	29 481.020	339.105, 341.477	14.3(7)		16.2
		2	30 619.440	336.156, 345.847	12.1(6)		13.1
$3d^9(^2D)4p$		3	29 668.918	336.958, 339.299	16.6(8)	17.2(7)[c]	17.0
$3d^9(^2D)4p$	$z\ ^3D°$	2	29 888.505	344.626, 350.085	15.7(8)		16.6
		1	30 912.838	342.371, 348.378	19.3(10)		21.2
$3d^8(^3F)4s4p(^3P°)$	$z\ ^3G°$	5	30 922.763	323.295	182(9)		137
		4	30 979.789	322.699, 337.200	305(15)		303
		3	31 786.210	323.466, 338.089	193(10)		154

(*Continued overleaf*)

Table 1. Continued

Configuration and Term		J	Level (cm^{-1})	Laser Wavelength in Air (nm)	Lifetime (ns) This Expt.	Lifetime (ns) Other LIF	Lifetime (ns) Critical Compilation[b]
$3d^9(^2D)4p$	$z\ ^1F^\circ$	3	31 031.042	324.306, 336.617	10.7(5)		12.0
$3d^9(^2D)4p$	$z\ ^1D^\circ$	2	31 441.665	332.026, 356.637	15.2(8)		15.9
$3d^8(^3F)4s4p(^3P^\circ)$	$^3F^\circ$	4	32 973.414	303.187, 305.083	15.4(8)	16.0(6)[c] 16.2(4)[d]	16.2
		3	33 112.368	303.794, 310.156	8.9(4)	9.1(4)[c] 9.1(4)[d]	9.6
		2	33 610.916	299.260, 305.432	8.0(4)	8.2(2)[c] 8.3(3)[d]	8.1
$3d^9(^2D)4p$	$z\ ^1P^\circ$	1	32 982.280	311.413, 338.058	6.5(3)	6.6(4)[c]	6.7
$3d^8(^3F)4s4p(^3P^\circ)$	$y\ ^3D^\circ$	3	33 500.854	298.413, 300.249	9.3(5)	9.6(3)[c] 10.5(5)[d]	9.6
		2	34 163.29	294.392, 300.363	10.4(5)	10.6(4)[c] 10.8(8)[d]	10.6
		1	34 408.574	298.165, 305.765	6.9(3)	6.9(3)[c] 7.5(6)[d]	6.9
$3d^8(^3F)4s4p(^3P^\circ)$	$z\ ^1G^\circ$	4	33 590.159	299.446, 309.912	96(5)	92(4)[c] 117(3)[d]	93
$3d^8(^3F)4s4p(^3P^\circ)$	$y\ ^1F^\circ$	3	35 639.148	282.130, 310.188	15.9(8)		17.9
$3d^8(^3F)4s4p(^3P^\circ)$	$y\ ^1D^\circ$	2	36 600.805	301.201, 433.164	6.4(3)	6.4(3)[c] 7.2(4)[d]	7.0
$3d^8(^3P)4s4p(^3P^\circ)$	$^5P^\circ$	3	40 361.254	247.688	231(12)		
		2	40 484.282	252.422, 269.650	405(20)		
$3d^8(^1D)4s4p(^3P^\circ)$	$^3F^\circ$	4	42 585.296	234.753, 242.333	44.8(22)		<45.5
		3	42 767.900	233.749, 238.659	13.0(7)		
		2	42 954.234	242.403, 240.185	26.8(13)		
$3d^8(^1D)4s4p(^3P^\circ)$	$^3D^\circ$	3	42 621.048	234.555, 242.123	4.2(2)		<4.5
		2	42 653.723	239.312, 241.931	29.1(15)		
		1	42 656.317	239.296, 247.207	28.7(14)		
$3d^8(^3F)4s4p(^1P^\circ)$	$^3G^\circ$	4	44 314.980	225.589, 232.580	2.9(2)		2.8
		3	44 565.10	225.355, 231.234	2.0(2)		<1.7
$3d^8(^3F)4s4p(^1P^\circ)$	$^3F^\circ$	4	43 258.792	231.096, 238.440	2.0(2)		
		3	45 281.152	221.777, 225.147	1.8(2)		1.8
		2	45 418.858	228.732, 231.398	1.8(2)		1.8
$3d^8(^1D)4s4p(^3P^\circ)$	$^3P^\circ$	1	43 464.019	242.366	108(5)		
		2	43 933.428	234.664, 239.639	23.3(12)		<18.2
$3d^8(^3F)4s4p(^1P^\circ)$		3	43 654.974	228.998, 230.077	3.4(2)		<3.5
$3d^8(^3P)4s4p(^3P^\circ)$	$^5D^\circ$	3	44 206.185	226.141, 230.735	42.5(21)		<33.2
$3d^8(^3P)4s4p(^3P^\circ)$		4	44 336.10	225.480, 232.465	3.5(2)		36.2
$3d^8(^3F)4s4p(^1P^\circ)$	$^3D^\circ$	2	44 475.158	225.813, 231.716	2.3(2)		2.3
		1	45 122.460	225.955, 230.297	2.1(2)		1.7
$3d^8(^3P)4s4p(^3P^\circ)$	$x\ ^3P^\circ$	2	46 522.965	215.831, 219.024	9.3(5)		9.1
		1	47 208.228	215.783, 219.738	7.5(4)		7.1
		0	47 686.625	217.448	7.3(4)		11.2
$3d^8(^3P)4s4p(^3P^\circ)$	$v\ ^3D^\circ$	3	47 030.148	213.493, 216.615	33.3(17)		
		2	47 139.392	216.104, 218.238	24.1(12)		<26.0
		1	47 424.830	214.780, 218.694	13.8(7)		
$3d^8(^3P)4s4p(^3P^\circ)$	$^5S^\circ$	2	47 328.85	212.140, 215.223	33.0(17)		<32.1
$3d^84s(^4F)5s$	$e\ ^5F$	5	48 466.530	440.155, 471.442	7.9(4)		8.1
		4	49 086.030	428.468, 445.905	8.6(4)		
		3	49 777.619	432.561	8.2(4)		<10.4
		2	50 346.477	422.171	8.1(4)		
$3d^9(^2D)5p$	$w\ ^3P^\circ$	2	48 735.308	205.992, 208.898	21.8(11)		
		1	49 403.42	206.020, 217.354	23.7(12)		26.3
		0	50 138.53	206.439	23.1(12)		25.0
$3d^9(^2D)4d$	$e\ ^3G$	5	49 158.529	427.139	11.3(6)		17.6
		4	49 174.811	444.148	11.5(6)		
$3d^84s(^4F)4d$	$e\ ^5H$	7	56 885.249	337.464	6.1(3)		
$3d^84s(^4F)4d$	$e\ ^5G$	6	56 954.167	336.681	6.4(3)		
$3d^84s(^4F)4d$	$f\ ^5F$	5	56 973.707	320.215, 336.459	6.3(3)		

[a]The numbers in parentheses are the uncertainties in the last digits of the entries.
[b]Ref. 7.
[c]Ref. 5
[d]Ref. 6.

Table 2. Comparison of Ni I Lifetimes Derived from Oscillator Strengths Measured by the Oxford Group[a] and Our Experimental Lifetimes

Upper Level	Energy (cm^{-1})	Oxford Contribution to Total Decay Rate (%)	Lifetime (ns)		Ratio
			Oxford[a]	This Expt.	
$z\ ^5F_4$	29 084	100	32.13	31.1	1.033
$z\ ^3P_2$	28 569	99	8.77	8.4	1.044
$z\ ^3P_1$	29 501	97	8.67	8.3	1.045
$z\ ^3F_3$	29 321	89	13.12	12.4	1.058
$z\ ^3F_4$	29 481	100	15.34	14.3	1.073
$z\ ^3F_2$	30 619	83	12.73	12.1	1.052
$z\ ^3D_2$	29 889	85	17.04	15.7	1.085
$z\ ^3G_5$	30 923	100	183.9	182	1.010
$z\ ^1F_3$	31 031	99	11.02	10.7	1.030
$z\ ^1D_2$	31 442	99	15.44	15.2	1.016
$^3F_3$	33 112	95	9.30	8.9	1.045
$^3F_2$	33 611	99	8.21	8.0	1.026
$y\ ^3D_3$	33 501	99	9.85	9.3	1.059
$y\ ^3D_1$	34 409	99+	7.09	6.9	1.028

[a]Ref. 4.

surements by LIF of Becker *et al.*[5] and our lifetimes are 1.0% and 2.7%, respectively.[5] The agreement with Ref. 6 is not quite so good as is typical for LIF lifetime measurements, but the agreement with the more recent measurements performed by Becker *et al.* is excellent. In all cases our measurements and those by Becker *et al.* agree to within 5%.

The critical compilation of atomic transition probabilities by Fuhr *et al.*[7] includes transition probabilities for essentially all of the decay channels of some selected Ni I levels. Lifetimes for these selected levels are derived by summing transition probabilities and are included in Table 1. Upper limits for a few additional lifetimes derived by summing transition probabilities are also included in Table 1. The critical compilation included most (80% or 90%) of the total transition probability for decay from these additional levels.

Typically the uncertainties in the transition probabilities in the critical compilation are ±25% for spectral lines from lower excited levels and range up to ±50% for lines from higher excited levels. These uncertainties are conservative. If we limit the comparison to upper levels for which the critical compilation has a complete set of transition probabilities and if we omit the $J = 4$ level at 44 336 cm^{-1}, then the agreement between our lifetimes and lifetimes from the critical compilation is extremely good. The average and root-mean-squared differences are +4.2% and 15.1%, respectively.

The most accurate (±0.7%) relative Ni I oscillator strengths (f values) are undoubtedly those measured recently by the Oxford group.[4] Blackwell *et al.*[4] used primarily Hanle effect measurements performed by Becker *et al.*[17] to normalize their relative absorption oscillator strengths. Blackwell *et al.*[4] estimate that the uncertainty in the absolute scale of their oscillator strengths is ±7%. Table 2 is a comparison between lifetimes derived from the Oxford oscillator strengths and our experimental lifetimes. The Oxford measurements include almost all (≥99%) of the transition probabilities from nine levels and include most (≥83%) from five additional levels. For 11 of the levels we use the assessment of completeness given by Blackwell *et al.*[4] The completeness of oscillator strengths from the $z\ ^1D_2$, $y\ ^3D_3$, and $y\ ^3D_1$ levels is assessed by comparison of the Oxford data with the recent critical compilation.[4,7] The ratio of these lifetimes is given in the last column. The average of all these ratios is 1.043, and the standard deviation is 0.021. The average of the ratios for which the Oxford contribution represents ≥99% is 1.035, and the standard deviation is 0.020. This comparison provides a new absolute scale for the Oxford f values; the $\log(gf)$'s should be increased by 0.015. The uncertainty in this $\log(gf)$ scale is no worse than ±0.009, which is the standard deviation of ratios (±2%), because the systematic uncertainties in our lifetimes are different for the long-lived and short-lived levels, as discussed previously. The +1% average difference between lifetime measurements by LIF of Becker *et al.* and our lifetimes shown in Table 1 also provides strong support for this revised absolute scale and for a ±2% total uncertainty on this scale.[5]

In summary, we report new radiative lifetime measurements for 66 levels in Ni I. Our measurements agree with earlier but less extensive LIF measurements; they also provide an improved absolute scale for the highly accurate measurements of relative oscillator strengths by the Oxford group. Finally, these lifetimes provide the absolute normalization needed to derive a comprehensive set of Ni I transition probabilities from Fourier transform spectra.

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